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SUPERCONDUCTING ELECTRONIC FILM STRUCTURES(U)

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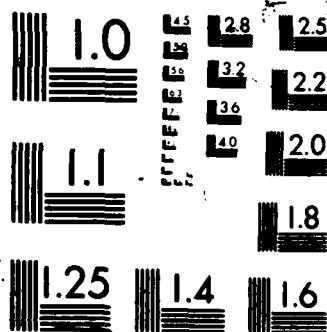
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Annual Report

AFOSR-TR 87-0314

SUPERCONDUCTING ELECTRONIC FILM STRUCTURES

A. I. Braginski and J. R. Gavaler
Cryogenic Technology and Electronics

January 20, 1987

Westinghouse Electric Corporation
Research and Development Center
Pittsburgh, Pennsylvania 15235

AFOSR Contract No. F49620-85-C-0043

Research sponsored by the Air Force
Office of Scientific Research, Air Force
Systems Command, United States Air Force

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SECURITY CLASSIFICATION OF THIS PAGE

REPORT DOCUMENTATION PAGE

1a. REPORT SECURITY CLASSIFICATION UNCLASSIFIED		1b. RESTRICTIVE MARKINGS										
2a. SECURITY CLASSIFICATION AUTHORITY		3. DISTRIBUTION/AVAILABILITY OF REPORT <i>Distribution Unlim. tcl</i>										
2b. DECLASSIFICATION/DOWNGRADING SCHEDULE												
4. PERFORMING ORGANIZATION REPORT NUMBER(S) 87-9C9-SUPER-R1		5. MONITORING ORGANIZATION REPORT NUMBER(S) AFOSR - TH- 87-0314										
6a. NAME OF PERFORMING ORGANIZATION Westinghouse Research and Development Center	6b. OFFICE SYMBOL <i>/</i>	7a. NAME OF MONITORING ORGANIZATION DCASMA Pittsburgh 53111A AFOSR										
6c. ADDRESS (City, State and ZIP Code) 1310 Beulah Road Pittsburgh, Pennsylvania 15235		7b. ADDRESS (City, State and ZIP Code) 1626 William S. Moorhead Federal Bldg. 1000 Liberty Avenue <i>Bellin, AFB</i> Pittsburgh, PA 15222 WASH. DC 2-332										
8a. NAME OF FUNDING/SPONSORING ORGANIZATION Air Force Office of Scientific Research	8b. OFFICE SYMBOL <i>NE</i>	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER F49620-85-C-0043										
9c. ADDRESS (City, State and ZIP Code) <i>Same as 7B</i>		10. SOURCE OF FUNDING NOS <table border="1"> <tr> <th>PROGRAM ELEMENT NO.</th> <th>PROJECT NO.</th> <th>TASK NO.</th> <th>WORK UNIT NO.</th> </tr> <tr> <td>61102F</td> <td>2306</td> <td>C1</td> <td></td> </tr> </table>		PROGRAM ELEMENT NO.	PROJECT NO.	TASK NO.	WORK UNIT NO.	61102F	2306	C1		
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61102F	2306	C1										
11. TITLE (Include Security Classification) SUPERCONDUCTING ELECTRONIC FILM STRUCTURES												
12. PERSONAL AUTHOR(S) A. I. Braginski and J. R. Gavaler												
13a. TYPE OF REPORT Annual	13b. TIME COVERED FROM 1/1/86 TO 12/31/86	14. DATE OF REPORT (Yr., Mo., Day) Jan. 20, 1987	15. PAGE COUNT 21									
16. SUPPLEMENTARY NOTATION												
17. COSAT CODES <table border="1"> <tr> <th>FIELD</th> <th>GROUP</th> <th>SUB. GR.</th> </tr> <tr> <td></td> <td></td> <td></td> </tr> <tr> <td></td> <td></td> <td></td> </tr> </table>	FIELD	GROUP	SUB. GR.							18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)		
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20. DISTRIBUTION/AVAILABILITY OF ABSTRACT UNCLASSIFIED/UNLIMITED <input checked="" type="checkbox"/> SAME AS RPT <input type="checkbox"/> DTIC USERS <input type="checkbox"/>		21. ABSTRACT SECURITY CLASSIFICATION UUU4										
22a. NAME OF RESPONSIBLE INDIVIDUAL DR. Weinstock		22b. TELEPHONE NUMBER <i>(Include Area Code)</i> 761-4931		22c. OFFICE SYMBOL NE								

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**A. I. Braginski and J. R. Gavaler
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1. Annual Report, Superconducting Electronic Film Structures.

January 1, 1986 to December 31, 1986

AFOSR Contract No. F49620-85-C-0043

A. I. Braginski and J. R. Gavaler.

2. ABSTRACT

Bulk samples of the newly discovered La-Sr-Cu-O superconductors have been made which have critical onset temperatures over 38 K. The greater effectiveness of impurities (oxygen) compared to epitaxy for the stabilization of stoichiometric A15 Nb-Ge was demonstrated. The positive effect of oxides on the formation of A15 Nb-Al was also shown. Experiments involving the deliberate addition of a contaminant (methane) during barrier deposition provided a major advance in the understanding of the processes required for producing high quality NbN-MgO-NbN junctions. A model for predicting the orientation of single crystal Nb and Nb-Sn films on sapphire was experimentally verified. Results were obtained which indicate that epitaxy is essential for NbN-MgO-NbN trilayer processing if the junction is to be operated above 4.2 K.

3. OBJECTIVES

The objectives of the Westinghouse-AFOSR Program are:

1. Investigate the low-temperature synthesis of high-critical-temperature superconducting films.
2. Grow epitaxially single-crystal superconducting films and coherent layered structures.
3. Characterize the near-boundary crystalline and phase perfection of superconducting layer surfaces and interfaces, mostly by in-situ methods.
4. Study tunneling into high-critical-temperature (T_c) superconducting films.
5. Study radio-frequency surface losses in high- T_c superconducting films.
6. Investigate artificial tunnel barriers.

4. ACCOMPLISHMENTS

4.1 Preamble

This five-year research program was initiated in January 1983. It is aimed at understanding and improving the superconducting and normal state properties of layered, epitaxial, thin film structures incorporating high-critical-temperature superconductors. Anticipated results are intended to form a material science base for a future technology of high operating temperature superconducting electronics. The initial work in this program was performed under a contract covering the period from January 1, 1983 to December 31, 1984. In 1985 the level of effort was augmented to include an additional objective (No. 6). During 1986 the level of effort was reduced somewhat and work toward one of the objectives (No. 5) was discontinued. Studies performed during the period from January 1, 1986 to December 31, 1986, on the five remaining objectives of this program are described in this report.

4.2 Low-Temperature Synthesis of High T_c Films

During the course of this program, all of the work on this objective has been focussed on the high- T_c A15 and B1 structure superconductors, for the obvious reason that these materials were the highest temperatures superconductors known. Very recently, however, a new class of compounds has been found which apparently exhibits superconducting critical temperatures more than twice as great as those of any of the B1 or A15 materials. These new materials have immediate relevance to the present program and therefore research on these compounds has been inaugurated. Results obtained thus far and also the present status of the studies of B1 and A15 structure compounds are presented below.

4.2.1 K_2NiF_4 Structure Compounds

In May of last year J. G. Bednorz and K. A. Muller of the IBM Zurich Laboratory in Switzerland published a paper on "Possible High T_c Superconductivity in the Ba-La-Cu-O System" [(Z. Phys. B- Condensed Matter 64, 189-193 (1986)]. They reported that compounds in the Ba-La-Cu-O system with the compositions $Ba_x La_{5-x} Cu_5 O_{5(3-y)}$ ($x = 1$ and 0.75 and $y > 0$) showed indications of superconducting behavior at temperatures greater than 30 K. Subsequent work by these authors and also now by many other researchers in at least a dozen different laboratories has confirmed the fact that these oxide compounds are indeed very high- T_c superconductors. The specific oxide phase which is superconducting has the K_2NiF_4 structure with the stoichiometry $La_{2-x} Ba_x Cu O_{4-y}$ where $x \sim 0.15$. This oxide tends to form with an oxygen deficiency, i.e. $y > 0$. Present preliminary indications are that the highest T_c 's are obtained when y is minimum. An isostructural phase in a related chemical system (La-Sr-Cu-O) has also been found to have a very high critical temperature by workers at the University of Tokyo and also at the ATT Bell Laboratories.

Work here at Westinghouse for the present program has initially concentrated on the La-Sr-Cu-O system. Bulk samples of $La_{1.8} Sr_{0.2} Cu O_{4-y}$ have been prepared which from X-ray analyses consist of an almost pure K_2NiF_4 phase. These samples have resistive onset T_c 's of ~38 K and zero resistance at ~35 K. Magnetization and Hall measurements have also been taken. From these measurements, a current density of $> 10^3 A/cm^2$ (between 0-6 Tesla at 4 K) and a carrier density of $10^{21}/cm^3$ are obtained. All of these results are included in a paper submitted to Appl. Phys. Letters.

From a technological point of view, the questions that undoubtedly will be asked are whether these new superconductors can be prepared in a useful form (i.e. as films or wires) and whether their properties (other than T_c) will lend themselves to device applications. To begin to answer such questions, exploratory work on the sputtering of thin films of these oxide superconductors has been started and will continue as part of Objective 1 of this program.

4.2.2 B1 Structure Compounds

In a paper published during 1986 in Vol. 32 of "Advances in Cryogenic Engineering," the understanding of the low temperature synthesis of B1 structure NbN films, as developed during the course of this program, is presented in detail. All of the more recent results on low temperature NbN film growth are consistent with the concepts and conclusions presented in that paper.

4.2.3 A15 Structure Compounds

It is by now well documented that Nb_3Ge , the highest- T_c A15 superconductor is unstable and therefore can be prepared only in metastable form at relatively low temperatures ($< 1000^\circ\text{C}$). Both epitaxy and impurities (oxygen) have been reported as successful means for stabilizing high- T_c Nb_3Ge . As described in previous reports, a new type of ultra-high vacuum deposition and in-situ analytical facility, referred to as the Superlattice Analytical and Deposition Facility (SDAF) has been implemented for use in this program. Because of its now proven capability for epitaxial film growth and because of its controlled high purity environment, this facility provides a unique tool for studying both epitaxy and impurities as possible means for stabilizing Nb_3Ge (or other) high- T_c superconductors. In a series of experiments completed during this reporting period, Nb-Ge single crystal films have been grown epitaxially on sapphire, Nb_3Ir , Nb_3Sn and cubic zirconia substrates. While average compositions of films were close to the ideal 3:1 stoichiometry, their T_c values were very low, about 6 K, with only a minor phase showing resistive onsets of up to 16 K (inductive 12 K). This indicated that the stabilization of the stoichiometric A15 phase had not occurred. To confirm this finding, the SDAF was modified slightly to allow the introduction of controlled levels of oxygen into the deposition chamber during the evaporation process. This allowed a direct observation of the effect of added oxygen impurities on the growth of the Nb_3Ge .

Oxygen was leaked into the UHV deposition chamber during deposition in such a way that the gas outlet was in the proximity of the

substrate block. Consequently, the measured O_2 pressures may have underestimated the partial pressure at the substrate surface. Experiments with oxygen were performed using sapphire substrates only. The highest pressures tested so far when co-evaporating Nb and Ge in the nearly stoichiometric proportion were 1 to 2×10^{-9} torr. At these O_2 pressures, the A15 phase critical temperatures increased dramatically, with typical resistive transitions between 19 and 15 K. Single crystal (100) RHEED and LEED patterns were still observable. From these results it is concluded that in an ultra high vacuum environment epitaxy alone cannot stabilize the high- T_c metastable A15 phase of Nb_3Ge and that the presence of oxygen is necessary.

It is also tentatively concluded that even higher oxygen pressures are necessary to approach 3/1 stoichiometry of the A15 phase and attain T_c 's above 20 K. It is not clear at this time whether, at these higher pressures, high quality (single crystal) epitaxy will still be possible.

Amorphous Nb-Ge films were deposited on (11-20) sapphire in UHV at room temperature and annealed at 880°C to test a published hypothesis that the only role of oxygen is to insure amorphicity in the initial stages of deposition. An additional rationale was to test for solid state epitaxy. The annealed deposits were polycrystalline with an indication of slight texturing. In the absence of oxygen additions these films were fully superconducting below 9 K. Addition of 1×10^{-9} torr of O_2 during the room temperature deposition increased the transitions to between 13.5 and 11 K independent of the film thickness. The XPS data obtained after annealing in UHV of 10^{-10} torr indicated that a significant part of niobium became oxidized to stable niobium oxides thus explaining the relatively low T_c 's in multiphase samples. It is concluded that these data disprove the need for amorphicity in the initial stage of Nb-Ge growth in order to stabilize the metastable high- T_c phase.

The initial data on Nb_3Ge films were reported at the 1986 March Meeting of the American Physical Society. A more complete version of

these results was reported in the Superconducting Materials Symposium at the Materials Research Society Meeting in December.

The study of the effect of oxygen (or oxide) impurities on the formation of another A15 material, Nb_3Al , was continued during this period. This study has involved the formation of Nb-Al diffusion couples, by sequential deposition of Nb and Al layers on sapphire or on niobium substrates, and their subsequent annealing at various temperatures. At 850°C Nb-Al bilayers deposited on oxide substrates reacted to form the A15 phase while similar layers on Nb substrates did not. These data were reported at the 1986 March Meeting of the APS.

A series of Nb-Al annealing experiments have since been completed which show that up to 880°C the A15 phase was formed only on the oxide substrates. Between 880 and 950°C the A15 phase was also obtained in couples on Nb substrates, however with T_c 's lower than those on the oxide substrates. The present results thus show unambiguously that in Nb-Al thin films, consisting initially of the bcc A2 phase, the presence of an oxide substrate reduces the activation (or nucleation) energy for the transformation of the A2 to the A15 phase. The results also show unambiguously that the A15 phase forms preferentially at grain boundaries and crystalline defects, where crystalline disorder and the presence of diffusing oxygen are most probable. In this respect it is important to note that the A15 phase nucleated within a single crystal A2 matrix is always polycrystalline. Evidently, the nuclei are not correlated crystallographically with the matrix structure since they form in areas involving defects.

The present results indicate that an interaction of a Nb-Al thin film with an oxide substrate facilitates the A15 phase formation. Such an interaction most likely involves a surface reaction, which can be initiated even at room temperature. In addition to the direct evidence obtained in this work (described in Section 4.4 of this report), the occurrences of nonequilibrium surface reactions between Nb or Al and Al_2O_3 , or other very stable oxides, are also inferred from interactions between oxide barriers and Nb counterelectrodes in superconducting tunnel junctions.

This phase of the study of Nb_3Al nucleation and growth is now completed. A scientific paper covering all of the results was published in the Journal of Applied Physics. The most significant results from this investigation are the data which for the first time unambiguously show that oxide impurities influence the formation of a relatively stable A15 phase (Nb_3Al). Previously this has been shown to be true only for the formation of the metastable Nb_3Ge A15 phase. This result thus lends credence to the concept that all A15 compounds form via some intermediate oxides analogous to how the prototype A15 phase (β -tungsten) is formed.

4.3 Epitaxial Growth of Superconducting Films

One of the critical objectives of the present work upon whose successful completion much of the program is dependent, is developing the capability of growing epitaxial superconducting films and barrier structures. When this program was undertaken one of the key but unproven starting assumptions was that in the highly controlled environment of the SDAF it would be possible to achieve this capability. Based on work to date, this assumption has been completely validated. The growth of epitaxial single-crystal films has become routine and is now a standard starting point for experiments performed under other tasks of the program.

The orientation of an epitaxial film is easy to predict if the film and substrate are isomorphic with similar lattice constants, such as $\text{Nb-Ge}(100)[010]$ (plane) [direction in the plane] grown on $\text{Nb}_3\text{Ir}(100)[010]$, or $\text{NbN}(110)[001]$ grown on $\text{MgO}(110)[001]$. However, current models of epitaxy cannot predict whether a film will grow as a single crystal, or its orientation if it does, if the film and substrate have dissimilar structures. A computer program has been written that aids in the identification of common atomic distances and surface symmetries. The input to the program is the atomic structure of a unit cell, and the Miller indices of a particular plane. The output is a view of a surface of the material cut parallel to the designated plane,

with the first layer of subsurface atoms also identified. For example, with the aid of LEED patterns, RHEED patterns, and a real-space picture of the $\alpha\text{-Al}_2\text{O}_3(1\bar{1}02)$ surface, one can identify that a spacing between oxygen ions in the alumina of 5.104 Å lies parallel to a spacing of 5.128 Å between tin ions in Nb_3Sn .

Single-crystal Nb_3Sn films have now been grown on three different orientations of sapphire. Films as thin as 10 nm had T_c 's of 16K. A qualitative understanding of the orientation relationships between sapphire and Nb_3Sn was reached by noting that Nb and Sn atoms appeared to occupy the Al sites on each of the sapphire surfaces. Comparison of the epitaxial growth of Nb_3Sn with epitaxial A15 compounds that have a different lattice constant, A15 Mo-Re, Nb_3Ge , and Nb_3Ir , gave an estimate of the maximum mismatch between Al-Al spacings and A15 atomic spacings that could still result in epitaxial growth. This work will be submitted to the Journal of Applied Physics. Results on epitaxial Nb-Ge were given in Section 4.2.3.

The qualitative model for the orientation of epitaxial films on sapphire has been found to apply to Nb films. The orientation of Nb on any face of sapphire can be predicted. The model was successfully tested by growing Nb with a predicted (311) orientation on $\alpha\text{-Al}_2\text{O}_3(11\bar{2}3)$.

Ultra-thin Nb films, < 5.0 nm thick, were studied in collaboration with S. Park of Stanford University to assess the effect of ion-beam cleaning the substrate on the interface cleanliness and structure of the deposit. It was found that ion cleaning removed a residual carbon contamination and some of the oxygen from the top monolayer of sapphire. The result was that strained Nb films, with the lattice constant of the sapphire, grew on the ion-cleaned surface and epitaxial films with the bulk lattice constant of Nb grew on substrates that had not been milled. This work is scheduled for publication in Physical Review B.

Superconductor/insulator/superconductor trilayers formed by single-crystal epitaxy have now been formed in a number of materials

systems: NbN-Al₂O₃, NbN-MgO, Nb₃Sn-Al₂O₃, and Nb-Al₂O₃. In the system of most technological interest, NbN-MgO, the gap voltages of tunnel junctions fabricated from the trilayers were > 5 mV. The high gap voltage is an indication of the value of the superconducting order parameter in the initial 4 nm deposit of the second NbN layer, which determines the operating temperature of the junction. Taken together with the results of other researchers in the field, epitaxial growth of tunnel barriers and top electrodes, by either single-crystal or polycrystalline epitaxy, appears to be essential to the operation of tunnel junctions above 4.2K. Data and considerations concerning these issues were presented at the 1986 Annual Meeting of the Materials Research Society.

The anisotropy of the upper critical field, H_{c2} , of NbN thin films was studied as part of a collaboration led by D. A. Rudman of MIT. A model was developed for films with columnar microstructures that was based on anisotropic conductivities and classical surface superconductivity, H_{c3} . Measurements of the angular dependence of single-crystal NbN films were an important test of the model since the conductivity could safely be assumed to be isotropic. The data for single crystals fit the standard H_{c3} model without the additional angular dependence that had been seen in columnar samples. This work will appear in the proceedings of the 1986 Applied Superconductivity Conference.

4.4 Characterization of Near-Surface Layers

The occurrence of non-equilibrium surface reactions between Nb and very stable oxides has been inferred from interactions between oxide barriers and Nb counterelectrodes in superconducting tunnel junctions. As discussed in Section 4.2.1 of this report, such reactions were also inferred from Nb-Al diffusion couple experiments. Direct experimental evidence was obtained from XPS studies of Nb/Al₂O₃ and Al/MgO interfaces. A few angstroms thick metal layers, evaporated at pressures of low to mid 10⁻¹¹ torr contained lower oxides (NbO) or defective

oxides (AlO_x where $x < 1.5$). Surfaces of thicker metal layers evaporated on top of these ultrathin films and analyzed in identical conditions were clean, without any signature of a lower or defective oxide. These results were published in the Journal of Applied Physics.

The properties of $\text{NbN}(100)$ and $\text{NbN}(111)$ single crystal film surfaces were studied at high temperatures to establish temperature limits for the formation of films and interfaces. Reconstructions of the two types of surfaces were observed by electron diffraction at 800°C for $\text{NbN}(111)$ and 900°C for $\text{NbN}(100)$. With the aid of XPS, T_c , and tunneling measurements, changes in surface structure were correlated with nitrogen loss from the surface in an ordered pattern. Models were developed for the structures of the surfaces before and after the nitrogen loss. A paper containing this work has been submitted to the Journal of Applied Physics.

Direct evidence has, for the first time, been obtained for the chemical interaction between a tunnel barrier and the initial deposit of a counterelectrode. Ultra-thin layers of NbN were deposited on MgO and found by XPS that < 1 monolayer of Nb suboxide formed at the interface. No evidence of the reduction of MgO was observed. Electron diffraction measurements indicated that the NbN layer grew with the 4% smaller lattice constant of the MgO for a thickness ≈ 1 nm. At a thickness of 4 nm, dislocations relieved the compressive strain.

The interface quality was independent of temperature up to 700°C . Tunnel junction counterelectrodes were deposited at that temperature and the junction measurements confirmed the XPS result that such high temperatures could be used without a destructive interdiffusion or reaction between the barrier and superconductor. Other experiments that led to the same conclusion were the annealing at 750°C of $\text{NbN}/\text{MgO}/\text{NbN}$ junctions (made at low temperatures) for the approximate time needed for counterelectrode deposition (15 min). The annealing resulted in no change in the tunneling properties. The survival of the barrier/superconductor interface during top electrode formation at high temperatures is desirable in the case of NbN where it

has been found by electron diffraction that increasing the deposition temperature from 150°C to 700°C improved the crystallinity of the first 4 nm deposited. However, it is a crucial intermediate step in the case of Al5 top electrodes since temperatures > 500°C are needed to form the Al5 compounds.

4.5 Tunneling into High-T_C Superconductors

The appearance of structure in the conductance of a superconductor/insulator/normal metal tunnel junction due to strong electron-phonon interactions in the superconductor has provided a means for measuring the phonon spectra of films. In collaboration with J. G. Adler of the University of Alberta, d^2I/dV^2 has been measured for NbN(111) single crystals and polycrystalline NbN. A controversy arose in 1985 between experimental groups at NRL and at Karlsruhe over the possible existence of a peak in the Eliashberg function, $\alpha^2F(\omega)$, at 45-50 meV. The present data did not exhibit such a peak, in agreement with theoretical calculations of the phonon structure.

The routine fabrication of low-leakage tunnel barriers that is now possible (Section 4.7) for NbN counterelectrodes, permitted the characterization by tunneling of a previously inaccessible region of the film - the initial deposit. By performing experiments with a fixed base electrode and barrier, tunneling into the top electrode was used to evaluate the effect of top electrode deposition temperature between 150° and 700°C. The biggest effect was measured for polycrystalline NbN/MgO/NbN junctions which had gap voltages increase over this range by 1.2 mV to values > 5 mV. The gap voltage of NbN(111)/MgO/NbN junctions increased by 0.6 mV. This work will be discussed in an invited presentation at the 1987 Annual Meeting of The Metallurgical Society of AIME.

4.6 Radio-frequency Surface Losses in High-T_C Superconducting Films

Work toward this objective has been suspended.

4.7 Artificial Tunnel Barriers

Major progress was made during the last year in the understanding of practical artificial barriers. The important issue that needed to be resolved has been the existence of "pinholes" in the barriers due to the inability of a thin overlayer to completely "wet" the superconductor surface. Documentation of this structure was published in September, 1986, in the Journal of Applied Physics. New deposition processes have been developed during the past year which minimize the deleterious effect of such pinholes in artificial oxide barriers on NbN. The results from these new processes can explain the large differences in the properties of junctions made in different laboratories under nominally the same conditions. The same processes and explanation have been found to apply to either single-crystal or polycrystalline junctions.

The essential ingredient in forming low-leakage all-NbN junctions has been found to be the introduction of some "contaminant" during the barrier sputtering process which helps to form a more stable native oxide in the pinholes. A stable niobium carboxide has been formed in the pinholes of sputtered MgO barriers by adding 1% CH_4 to the Ar sputter gas, or by stopping at the midpoint of barrier deposition, exposing the barrier to air, and then completing the deposition. XPS data show that these processes incorporated carbon into niobium oxide and the total thickness of this niobium oxide became much greater than for oxides deposited in pure Ar. It is believed that by using slow deposition rates in the oil diffusion pumped vacuum chambers which are used by some workers, similar stable niobium carbo-oxides can be and are in fact (unintentionally) produced. The resulting junctions made in such relatively low-purity environments are then greatly superior to those prepared in a contamination-free UHV system. These new data on artificial tunnel barriers were presented at the 1986 Annual Meeting of the Materials Research Society.

A trilayer of polycrystalline NbN/oxidized Al/NbN, the same material used for 100-junction series arrays (R. D. Blaugh et al.,

proceedings 1986 Applied Superconductivity Conference), was prepared for cross-sectional transmission electron microscopy (XTEM) and the first real-space images of an artificial tunnel barrier were obtained. One of these micrographs will be published in the proceedings of the 1986 ASC. Concurrent with the ASC, the first XTEM micrographs of an all-NbN junction with a native oxide were published by the Hitachi group (J. Appl. Phys. 60, 2187, 1986). The micrographs of junctions with artificial barriers and those with native oxide are virtually indistinguishable. Both showed surface roughness of the NbN base of ~ 5 nm and obvious non-uniformity of the barrier thickness. The Hitachi conclusion was that it may be necessary to use a single-crystal NbN base to obtain barrier uniformity.

XTEM has since been used to look at a single-crystal NbN(100)/MgO/NbN trilayer and a NbN(111) film. In both cases, the base NbN roughness was reduced over the polycrystalline trilayers, and barrier uniformity was clearly improved. These micrographs will be included in the proceedings of the 1987 TMS-AIME Annual Meeting.

5. PUBLICATIONS

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*Submitted for publication in 1985.

6. PERSONNEL

A. I. Braginski }
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7. COUPLING ACTIVITIES*

1. "Properties of Epitaxial Nb-Ge Films on Nb₃Ir and Nb₃Sn Single Crystals," M. A. Janocko, A. I. Braginski, J. R. Gavaler, and J. Talvacchio, contributed talk to the March 1986 American Physical Society Meeting.
2. "Formation of the A15 Phase in Nb-Al Diffusion Couples," J. R. Gavaler, A. I. Braginski, and K. Schulze, contributed talk to the March 1986 American Physical Society Meeting.
3. "Epitaxial Growth of NbN/MgO/NbN Multilayers for Tunnel Junctions," J. Talvacchio, A. I. Braginski, M. A. Janocko, and J. R. Gavaler, contributed talk to the March 1986 American Physical Society Meeting.
4. "UHV Preparation and Analysis of Layered Superconducting Thin Film Structures," A. I. Braginski, Invited Seminar at Yale University, New Haven, April 18, 1986.
5. "Formation of Metastable A15 and B1 Superconducting Phases," J. R. Gavaler, Invited Presentation, 1986 Annual Meeting of the Materials Research Society, Boston, Mass.
6. "Epitaxial Superconductor/Insulator Layered Structures," J. Talvacchio, Invited Presentation, 1986 Annual Meeting of the Materials Research Society, Boston, Mass.
7. "Epitaxial Tunneling Film Structures," A. I. Braginski, Seminar at Stanford University, December 15, 1986.

*Speaker's name is underlined.

8. PATENTS AND INVENTIONS

Patent Disclosure No. RES86-222: "Tunnel Junctions with High- T_c Electrodes of an Al₅ Compound Fabricated by Diffusion," by J. R. Gavaler and A. I. Braginski.

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5 - 87

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